Spin orientation in an ultrathin CoO/PtFe double-layer with perpendicular exchange coupling

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Abstract

We studied by soft x-ray absorption spectroscopy the magnetization axis in an 4nm thin CoO (111) layer exchange-coupled to an ultra thin $L1_0$ PtFe layer with perpendicular magnetic anisotropy. The angular dependence of the linear magnetic dichroism at 10K and the relative variations of the spectral features provide a full description of the spin orientation in this antiferromagnetic layer. The spins are found in the film plane, pointing along the 110 direction. This results is discussed in relation to the film strain and preferential occupation of t_{2g} orbitals.The strong orthogonal coupling between Co and Fe spins should be at the origin of the robustness of the exchange bias effect foun in this bilayer system.

Keywords: x-ray absorption spectrocopy, x-ray linear dichroism, multiplets, antiferromagnetic

1. Introduction

Antiferromagnetic/ferromagnetic (AFM/FM) bilayers with controllable perpendicular magnetization are of a great interest for potential logic devices with high density, thermal stability and low critical current for current-induced magnetization [1, 2]. To have an adequate description of AFM/FM couplings, crystallographic order, epitaxial strain, spin orientation and competing anysotropies are essential parameters.

The PtFe alloy in the chemically-ordered $L1_0$ phase has one of the strongest magnetocrystalline anisotropy energy. The $L1_0$ phase, formed by alternating Fe and Pt atomic planes along the c-axis of the tetragonal structure, leads to an enhanced uniaxial anisotropy along the ordering axis [3] even in the thinnest limit of a few monolayers (ML) [4, 5]. $L1_0$ PtFe(001) layer with tetragonal axis perpendicular to the surface provides then an ideal out-of-plane spin network with enhanced perpendicular magnetic anisotropy to study the exchange-coupling properties with thin AFM layers. Among them CoO films are specially relevant for spintronic devices. Bulk CoO has a Néel temperature (T_N) of 293 K, close to room temperature (RT), and a magnetic moment of 3.98 μ_B [6, 7], with a large orbital contribution. In the paramagnetic phase, bulk CoO crystallizes in the rocksalt structure with alternate Co and O (111)-planes (fig.1). Below T_N, AFM ordering goes along with a monoclinic distortion [6, 7]. The bulk CoO AFM structure is described as a stacking of uncompensated FM hexagonal planes of Co^{2+} $3d^7$ ions coupled antiferromagnetically along the trigonal elongation (fig.1). In thin CoO layers grown on different substrates, polarization dependent soft x-ray absorption spectroscopy shows that the strain induced by epitaxy leads to significant modifications in the magnitude and orientation of the magnetic moments [8], confirming the important sensitivity to magnetostriction. These results motivate our combined investigation of structural distortion and spin orientation in an ultrathin CoO/PtFe double-layer with perpendicular exchange coupling.

2. Experiments

The films have been grown by molecular beam epitaxy and studied *in situ* by grazing incidence x-ray diffraction (GI-XRD) at the French CRG BM32 beamline at the European Synchrotron Radiation Facility (ESRF, France). The Pt-terminated $L1_0$ PtFe(001) thin

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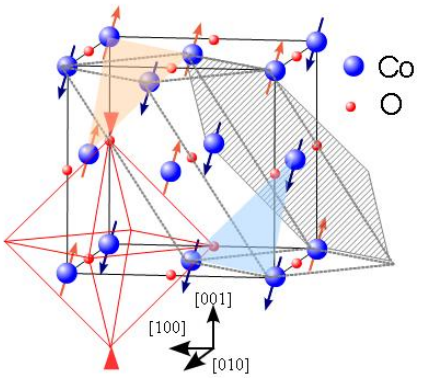

Figure 1: bulk CoO structure showing the Co(111) FM sheets coupled antiferromagnetically along the [111] direction. The hached surface corresponds to the hexagon lying on the FePt layer in the CoO/PtFe bilayer. An CoO6 octahedron is shown

layer grown by thermal deposition of about 3 MLs of Fe on an ultrahigh vacuum cleaned Pt(001) substrate [9] hold at 600 K is in coherent epitaxy on Pt(001). A CoO layer with 4nm thickness was grown by reactive molecular beam epitaxy on the ultrathin PtFe(001) layer hold at 523 K [10]. GI-XRD shows that the growth by reactive molecular beam epitaxy of the cobalt oxide on a Pt-terminated PtFe/Pt(001) surface gives rise to an hexagonal (111)-like surface. The strain imposed by the substrate on the CoO layer leads to a slight monoclinic distortion at room temperature. The monoclinic distortion can be described as a tetragonal distortion with c/a=1.008, with an additional slight trigonal distortion along the rocksalt [111] direction. The [001] axis of the tetragonal distortion is not perpendicular to the surface, but tilted by θ_0=54.74° in relation to the normal surface. The growth proceeds in fourfold equivalent well-crystallized domains, so that the [001] axis is fourfold degenerated. The detailled growth procedure and x-ray diffraction study will be described elsewhere [10]. The double layer shows perpendicular magnetic anisotropy and an exchange bias shift of 800 Oe at 10 K [11].

The orientation of the spin moments in the CoO layer at low temperature (T ≈ 10 K) was investigated *ex situ* by soft x-ray absorption spectroscopy (XAS) using linear dichroism at Co $L_{2,3}$ edges, at the PGM beamline of the Laboratorio Nacional de Luz Sincrotron (LNLS, Brazil). The spectral resolution was E/ΔE=6000 and the degree of linear polarization close to 100%. The sample was cooled down with an applied magnetic field of 5 kOe perpendicular to the surface plane. The magnetic field was then set to zero and the spectra were collected using the total electron yield. The sample was allowed to rotate about a vertical axis, with the polar angle (θ)

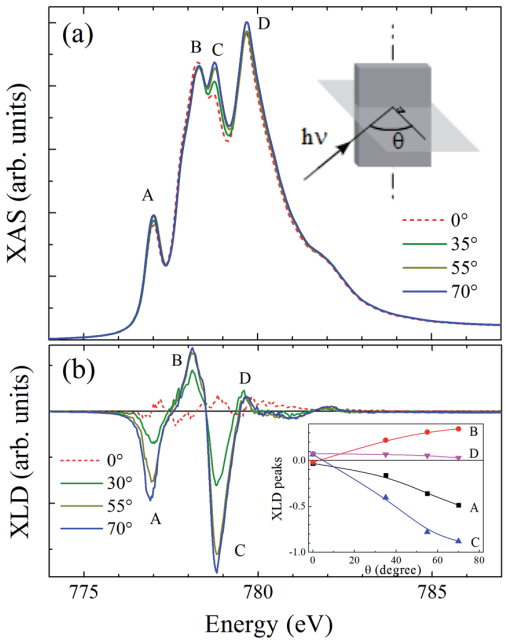

Figure 2: (a) Co L_3 edge XAS as function of θ. Iinset: experimental geometry; (b) XLD as function of θ. Inset : angular variation of the intensity of the main XMLD features

defined as the angle between the surface normal and the x-ray propagation (fig.2-a, inset). In this experimental geometry the variation of the escape length of the electrons as a function of θ should be corrected in order to recover the real x-ray absorption signal. This has been systematically corrected using the standard procedure for electron yield saturation effects [12]. To check this correction, for each θ, the x-ray absorption spectrum collected with vertical polarization was used as reference. At each angle, the x-ray linear dichroism (XLD) is obtained by two ways : (1) from the difference between the absorption with horizontal and vertical x-ray polarizations and (2) with horizontal x-ray polarization, from the difference between the absorption at θ and at θ =0°. The two methods give the same signals, within the accuracy of the measurements.

3. Results

Figure 2-a shows the Co L_3 edge XAS spectra at 10 K for four θ values from θ=0° (polarization vector in the surface) to θ=70° (polarization vector towards the surface normal). The spectra, normalized far from $L_{2,3}$ edges, show a clear linear dichroism. Four main features (labelled A to D) are observed in the XAS and XLD signals. They correspond to transitions towards orbitals of

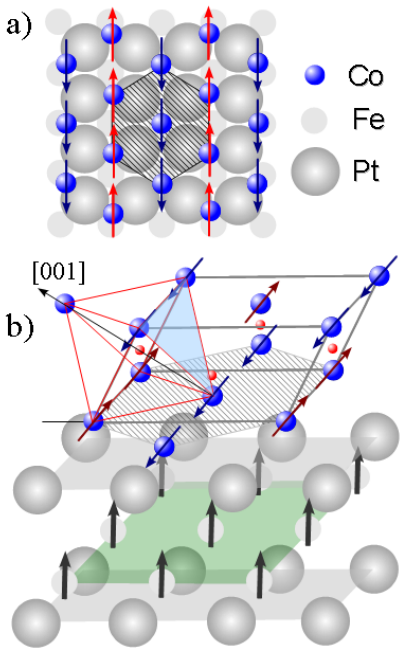

Figure 3: Sketches of the spin orientation in the CoO/PtFe bilayer (a) top view; (b) perspective. The [001] axis of tetragonal elongation is indicated in the CoO6 octahedron.

different symmetries and then show distinct variations as a function of θ (figure 2-b, inset). Taking the θ=0° spectrum as reference, the feature D at higher energy is almost constant, feature B increases with θ, while A and C decrease.

Magnetic and non-magnetic effects mixed by the local crystal field symmetry contribute to the polarization contrast. However, we have observed that the angular dependence dramatically decreases when the temperature increases and almost vanishes above 300 K, when the magnetic contribution to the anisotropy has vanished [11]. The XLD signal at 10 K, then, measures essentially the magnetic dichroism, i.e. the charge anisotropy associated to the magnetic moment anisotropy through the spin-orbit coupling [13, 14, 8]. Relatively to the magnetic axis, this anisotropy due to local exchange fields and spin-orbit coupling writes: $I_{XMLD} \propto |\vec{m}.\vec{E}|^2$, with $\vec{E}$ the electric field polarization and $\vec{m}$ along the magnetic axis. A $cos^2\theta$ dependence is then expected for the magnetism-sensitive transitions. The feature C shows the largest dichroism and fits well with a $cos^2\theta$ function, with a minimum at theta=90°. From multiplet calculations it has been demonstrated that the situation when the polarization vector is perpendicular to the magnetic axis, $\vec{m} \perp \vec{E}$, corresponds to C minimum and B maximum [13, 14]. This is obtained for θ=90°, within an accuracy of a few degrees. We can then conclude that the Co spins are essentially parallel to the surface plane. As the FePt spin axis is perpendicular to the layer, the coupling between Co and Fe spins at the interface is orthogonal (fig.3).

Additional information on the orientation of the spins within the plane can be drawn from the relative variation of the different features. For the same relative orientations of $\vec{E}$ and $\vec{m}$, multiplet calculations show quite different XAS and XLD features when the spin axis $\vec{m}$ are along <110> or along <100> direction [13]. In the first case A and C features have the same variation with θ, in the second their variations are opposite. The experimental data show that A and C vary in the same way (figure 2-b, inset). We deduce that the Co spins are pointing along the <110> direction.

4. Discussion

The relationship between the orientation of the magnetic moments and the sign of the crystal field effect has been demonstrated by Csiszar and coworkers [8]. In the case of CoO(001) on Ag(100), the strain induces a small tetragonal distortion with c/a >1. In the CoO_6 octahedron the elongation of the distances along the c axis goes along with a splitting of the t_{2g} orbitals. The partial filling of the Co^{2+} t_{2g} orbitals in this geometry is recalled in figure 4-a. The low lying t_{2g} d_{xz} and d_{yz} orbitals are filled and the t_{2g} hole will occupy the higher d_{xy} orbital. We should note that in our film the tetragonal distortion is weak (c/a <1.008) and that its influence on the orbital occupancy may be called into question. The integrated intensity of the XAS spectra may provide a direct experimental verification of the orbital occupation [8]. We have calculated the integral I_θ over the entire $L_{2,3}$ spectral region (775eV to 801eV) for the different angles θ and plotted in figure 4-b the ratio $(I_\theta\text{-}I_0)/I_0$. The increase of this ratio gives clear evidence of the increase of accessible t_{2g} holes as θ increases. In a simple experimental geometry of CoO(001) on Ag(001), where the film plane would also be the d_{xy} orbital plane, we could straightforward conclude about the preferential orbital occupancy. In the case of CoO(111) on PtFe/Pt(001), however, the normal of the planes containing d_{xy} orbitals are tilted by an angle of θ_0=54.74° in relation the surface normal (see fig. 3) and the growth proceeds in fourfold equivalent domains. Instead of a simple $cos^2(\theta)$ angular dependence, the intensity $I_{xy}(\theta)$ of the transition towards d_{xy} orbitals should be geometrically derived and averaged over the four domains. We found that $I_{xy} \propto cos^2(\theta\text{-}\ \theta_0) + cos^2(\theta\text{+}\ \theta_0)$. This function shows a maximum for θ=90° and minimum at θ=0°. The experi-

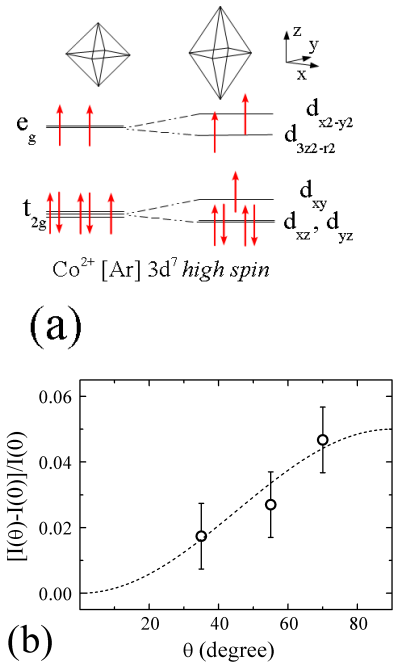

Figure 4: (a) Shematic figure of the 3d orbital occupancy for Co^{2+} in octahedral environment. For the axial elongation of the point charge octahedron along z, the degeneracy of the t_{2g} orbitals is lifted and the lowest level unpaired electron lies in the dxy orbital; (b) experimental angular variation of the integrated intensity of the XAS spectra (dots). The dotted line corresponds to the calculated $I_{xy}(\theta)$ function.

mental results (fig.4-b) confirm then a preferential localization of the t_{2g} holes with d_{xy} symmetry. With the spin-orbit coupling tending to align the spin in this same plane, we can expect the spin along the common direction of the d_{xy} planes and the surface plane $(\bar{1}\bar{1}1)$, i.e. the Co spins must be along the $[1\bar{1}0]$ direction (fig.3). This is in full agreement with the conclusions drawn from the study of the multiplet features.

Co spins are parallel to the surface plane, so that the coupling between Co and Fe spins at the interface is at 90 degrees. This 90 degrees coupling has been reported for the (in-plane anisotropy) CoO/Fe interface on Ag(001) [14], where the AFM interface is formed by compensated Co spins. Such orthogonal coupling is, indeed, the most stable configuration when the exchange field of a FM layer interacts with a completly compensated AFM layer. Both the good crystallinity of the interface and the robust orthogonal coupling should be at the origin of the robust exchange bias found in this bilayer system [11].

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